Progress in applying the FKK multistep reaction theory to intermediate-energy data evaluation

M.B. Chadwick[1], P.G. Young[2] and F.S. Dietrich[1]

[1] *University of California, Nuclear Data Group, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA*

(2) *University of California, Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

Recent developments to the physics modeling in the FKK-GNASH code system are reviewed. We describe modifications to include a linking of multistep direct and multistep compound processes, which are important when the incident energy is less than about 30 MeV. A model for multiple preequilibrium emission is given, and compared with experimental measurements of proton reactions on ^{90}Zr up to 160 MeV. We also give some preliminary observations on FKK calculations using DWBA matrix elements which are modified to include an inverse S-matrix factor – this addresses a long-standing controversy concerning the appropriate boundary-condition for multistep processes.

We describe the application of the FKK-GNASH code to a range of nuclear data applications, including intermediate energy reactions of importance in the accelerator transmutation of waste, and fast neutron and proton cancer radiation treatment. We outline areas where further work is needed for the accurate modeling of nuclear reactions using the FKK theory.

I. INTRODUCTION

The FKK-GNASH code is a version of GNASH [1] which uses the Feshbach-Kerman-Koonin (FKK) theory [2] for preequilibrium emission of nucleons. The Hauser-Feshbach theory is used for equilibrium decay, in an open-ended sequence of reaction chains with full conservation of angular momentum. Transmission coefficients for particles are obtained from an optical model, and for gamma rays from a generalized Lorentzian giant resonance model.

The FKK-GNASH code was developed with a view towards improved modeling capabilities up to 200 MeV. It has been used in a number of analyses in the 14-26 MeV range [3–6], and was extended to analyze higher energy reactions up to the pion threshold [7–9]. In Ref. [10] we described recent developments to the GNASH and FKK-GNASH code systems. In considering reactions at higher energies it became clear that certain extensions to the FKK theory had to be made. In particular, inclusion of multiple preequilibrium processes was found to be essential when calculating emission spectra while simultaneously satisfying flux conservation [7, 8]. We had already observed [11] the need for including multiple preequilibrium in analyses of $(n, xn\gamma)$ excitation functions up to 200 MeV, and analyses of emission spectra provided further evidence for their importance. Other improvements that we have made in higher-energy modeling include a description of preequilibrium spin effects [4], and a theory linking the direct and compound multistep chains [3, 5]. In this paper we give an account of some of these developments.

According to the FKK theory, a nuclear reaction takes place in a series of stages corresponding to the interaction of the incident particle with nucleons in the target nucleus. At each stage, nucleons are excited to higher states and may be emitted; these are the preequilibrium reactions. The theory distinguishes between two types of interaction, the P-chain in which the projectile remains always in the continuum and the Q-chain where all the nucleons are bound after the initial interaction. The P-chain dominates at higher energies and gives cross sections peaked in the forward direction, while the Q-chain becomes important at low energies and gives cross sections symmetric about 90 degrees characteristic of compound nucleus emission. Emission from the P-chain is referred to as multistep direct (MSD) and emission from the Q-chain as multistep compound (MSC).

At incident energies above 10-20 MeV the MSD process dominates, especially for the higher outgoing energies. The cross sections for such reactions have been extensively compared with the FKK theory, generally with good results. For a recent review see Ref. [12]. At lower energies both the MSC and MSD processes contribute. The MSC formalism is given in the review of Bonetti *et al.* [13]. These analyses show the usefulness of the FKK theory in understanding the cross sections of nuclear reactions over a wide range of energies. The FKK-GNASH code includes both MSD and MSC calculations and calculates the whole emission spectrum in a consistent manner. Details of the theoretical formalism used can be found in Refs. [3, 8].

In Sec. II we describe recent developments in FKK analyses that can be important for modeling nuclear reactions up to 200 MeV: the development of a multistep scattering theory that links MSD and MSC processes; the inclusion of multiple preequilibrium emission; and investigations into the appropriate boundary conditions for matrix elements in multistep processes. In Sec. III we discuss the FKK-GNASH code's use in a number of applications. A summary of some of the outstanding questions that still need to be solved in FKK analyses is given in Sec.IV.

II. RECENT THEORY DEVELOPMENTS

A. Linking of MSD and MSC processes

Recent FKK analyses by Chadwick *et al.* [3, 14] and Marcinkowski *et al.* [15] have pointed to the importance of transitions from the MSD P-chain to the MSC Q-chain. These mechanisms were ignored in the original FKK paper, which assumed no crossover transitions after the initial separation into P and Q space. Evidence for $P \rightarrow Q$ transitions comes from two considerations:(1) Analyses of the partitioning between MSD and MSC emission; and (2) Unitarity. The angular shape of the emission spectra in the preequilibrium emission regime determines the emission from the early MSC stages, and consequently the production of the early MSC stages. However, unitarity dictates that flux not lost to MSC or MSD preequilibrium emission must decay by compound nucleus emission. From such considerations it was concluded that a certain amount of reaction flux must bypass the initial Q-chain stages, entering the Q-chain after a number of collisions in P-space. It is not surprising that the Q-chain should be populated in this manner, since the probability of all particle excitations being bound will increase after more nucleon-nucleon collisions degrade the incident particle energy.

These works have postulated that the probability of flux entering a given Q-stage, with p particles and h holes and energy E, is given by the phase space ratio $\omega^B(p, h, E)/\omega(p, h, E)$, where the superscript "B" implies that only bound excitations are included. This prescription was checked by Chadwick and Young [3] for the initial flux entering the Q-chain, and was found to account for experimental data reasonably well. Also, independent investigations by Sato and Yoshida [16], on the imaginary optical potential in preequilibrium analyses partly confirmed this partitioning.

A full theoretical description of the coupling of MSD and MSC chains has recently been developed by Arbanas *et al.* [5], who removed the FKK assumptions which prevented $P \rightarrow Q$ transitions. They showed that in general there can be a number of P-space scatterings before, or after, scatterings in Q-space, and presented expressions for determining these processes. The attractive convolution structure of the MSD theory also appeared in the expressions for linked multistep scatterings in P and Q space.

The main physical consequence of these processes is that flux passing through preequilibrium stages can bypass the initial MSC Q-chain stages. Since it is predominantly these initial stages that give the high-energy MSC emission, this results in a reduced MSC component, and increased compound nucleus emission.

B. Multiple Preequilibrium Emission

Recent work [8] has shown that FKK analyses at incident energies above about 50 MeV should include multiple preequilibrium mechanisms, where more than one particle can be emitted from a preequilibrium stage. These processes were accounted for using a model which is very easy to implement computationally, and which does not require any additional DWBA matrix elements to be determined. It was assumed that any particles left excited after primary preequilibrium can also immediately escape with transmission-coefficient probability.

It should be noted that the FKK-GNASH calculations [7] submitted to the NEA intermediate energy code intercomparison [17] used an earlier exciton-model algorithm for the multiple preequilibrium [1, 7]. Our new model, described in detail in Ref. [8], provides a more consistent way to describe these processes in FKK analyses.

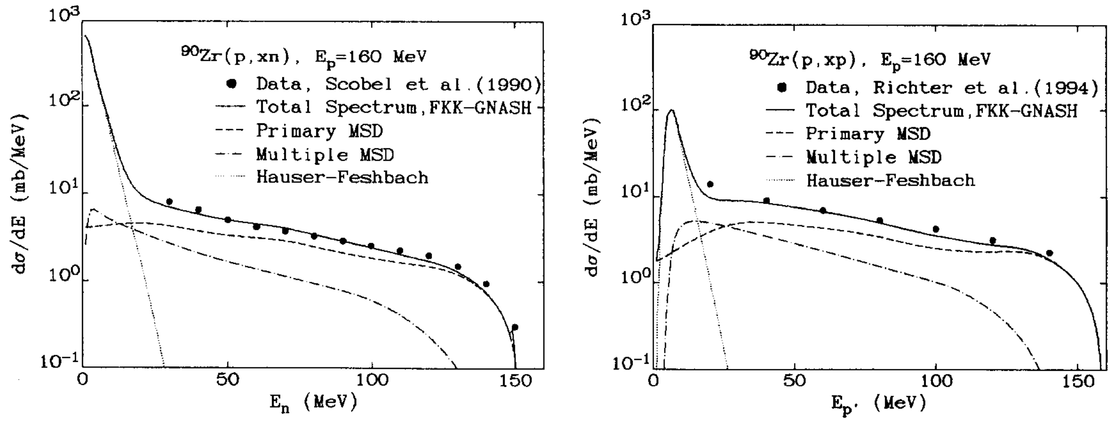

Fig. 1. Primary and multiple MSD, and Hauser-Feshbach, compared with data for the 160 MeV reactions ^{90}Zr(p, xn) and ^{90}Zr(p, xp).

In Fig. 1 we show comparisons between theoretical predictions and experimental measurements [18, 19] of angle-integrated neutron and proton emission spectra for proton-induced reactions on zirconium at 160 MeV. The full curves represent calculations including all reaction mechanisms contributing to the inclusive spectra. The contribution from primary MSD is shown by the dashed line, multiple MSD by the dashed-dot line, and Hauser-Feshbach equilibrium decay by the dotted line. It is evident that the calculations account for the measurements well, for both neutron and proton emission. The importance of multiple preequilibrium emission can be clearly seen for all emission energies except the very highest, and at the lower emission energies this mechanism accounts for much of the data. Furthermore, it was shown in Ref. [8] that if the FKK theory is used to account for emission spectra without including multiple preequilibrium, it is likely that unitarity is violated.

In Ref. [11] we also demonstrated the importance of including multiple preequilibrium emission when describing excitation functions in ^{208}Pb$(n, xn\gamma)$ reactions, for incident neutron energies up to 200 MeV, and for x=1-9. Including multiple preequilibrium greatly enhances the production of the residual nuclei ^{207,206}Pb (i.e. x=2,3), since the fast preequilibrium particles carry off much of the available incident energy. Likewise, production of residuals such as ^{200}Pb (i.e. x=9) is then strongly reduced since there is less energy available for many sequential compound–nucleus decays. Without including multiple preequilibrium emission the calculations would underpredict the data by orders of magnitude in some cases.

C. Boundary conditions for multistep direct reactions

There has been a long and so far unsettled controversy regarding the boundary conditions to be used on the intermediate-state wave functions in the FKK MSD theory. Up to the present nearly all calculations use normal DWBA matrix elements, as advocated by Feshbach [20], which conveniently allows MSD to be calculated by convoluting 1-step cross sections. However, Feshbach's arguments for this procedure have not been universally accepted. An alternative procedure, in which the boundary conditions are those naturally appearing in the complete set of states inserted in the evaluation of the intermediate-state optical Green's function, has been advocated by Kawai and others, and noted recently in Koning and Akkermans' derivation [21]. In order to investigate this issue, we have performed FKK calculations using a version of DWUCK4 modified to calculate the modified DWBA matrix elements (MDW) (which include an inverse S-matrix factor) required in the second approach. We find that this procedure yields multistep contributions that are greatly enhanced (a factor of 10 enhancement for the 2-step MSD is typical). We have the following preliminary observations concerning calculations based on these two approaches:

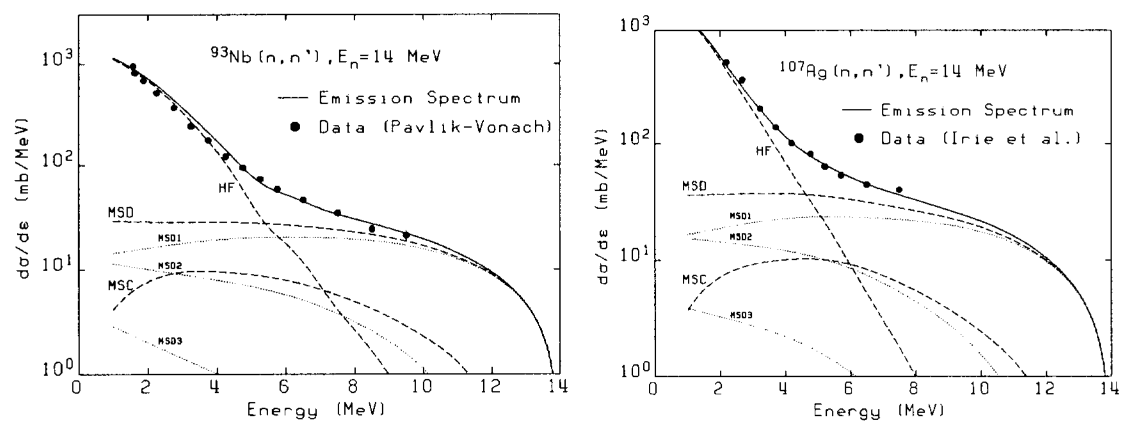

Fig. 2. FKK and Hauser Feshbach analyses of the 14 MeV ^{93}Nb(n, xn) and ^{107}Ag(n, xn) reactions, using the MDW approach. The MSD, MSC and Hauser-Feshbach contributions are shown by the dashed lines. The various MSD steps that sum to the total MSD spectrum are indicated by the dotted lines.

- In the 10-30 MeV incident energy range, both procedures describe data well. While the multistep contributions are bigger using MDW, they are still small compared to the 1-step process. However, at 14 MeV, we find that the magnitude of multistep contributions from the exciton model is similar to that from the

MDW calculations. Figure 2 shows results for 14-MeV inelastic scattering on ^{93}Nb and ^{107}Ag. For both these cases we used a residual interaction strength of V_0=36 MeV, and the standard input parameters as used by Chadwick and Young [3] with the Wilmore-Hodgson neutron potential. Agreement is satisfactory for both targets using the MDW procedure. This result differs from that of [22], in which the calculated two-step contribution for the ^{107}Ag reaction is much larger than ours; the reason for the difference is not understood, but may possibly be related to different treatment of the level densities and form factors.

- At higher energies we find MDW multistep contributions which are too large, giving spectra that are of the wrong shape. However, it is possible that this is due to inadequacies in other assumptions, e.g. in the model used for single-particle level densities at high excitations, rather than the matrix elements themselves. As an example, we show in Fig. 3 the calculated spectrum compared with data [23] when the MDW approach is used for the 80 MeV ^{90}Zr(p,n) reaction, including five preequilibrium steps. A value for the residual interaction strength of V_0=9 MeV was chosen to account for the highest emission energies where 1-step dominates. At the lower energies the multistep contributions are too large and result in an incorrect spectrum shape and a break-down of flux conservation.

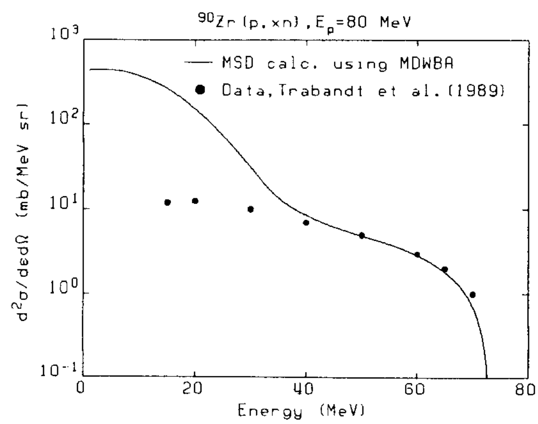

Fig. 3. Calculated MSD using the MDW approach for 80-MeV ^{90}Zr(p,n) reaction.

- The theory of Arbanas *et al.* [5] only yields significant $P \to Q$ flux when the MDW theory is used. As discussed in Sec. IIA, there are reasons to believe that such $P \to Q$ processes are significant

In short, we have contradictory results which still need to be understood. In practice the theory with the normal DWBA matrix elements fits the data reasonably well and will undoubtedly continue to be used as a computational tool until the high-energy problems with the MDW approach are resolved. Further work leading to understanding these issues is extremely important, since basing nuclear data calculations on a theory without an agreed implementation is fundamentally unsatisfactory.

III. NUCLEAR DATA EVALUATION

Over the last two years there have been considerable developments in nuclear data evaluation using the FKK theory, and here we briefly summarize this progress.

In order to use the FKK theory in applications, it is essential that the sensitivity of the calculations to different input parameters is understood. Watanabe [24] has shown that the value of the extracted residual interaction strength V_0 (the only free parameter in the theory) is rather sensitive to the choice of optical model, spin cut-off, and level density parameters, though the shape of the calculated spectra is not. Since it is desirable to find the systematical energy dependence of V_0 for predictive purposes, it is clear that a "standard" set of input parameters should be established for the analysis of different data sets. In this way the systematic behavior of V_0 can then be used with confidence to evaluate reactions where no measurements exist.

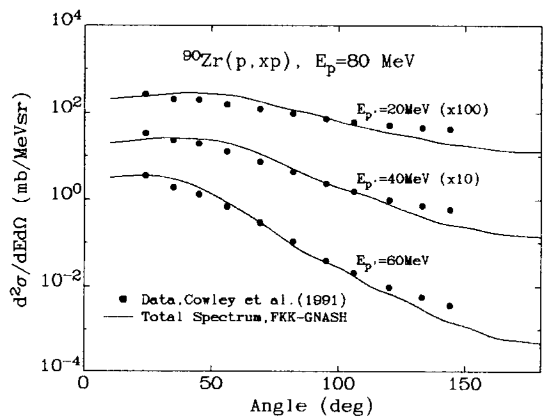

Fig. 4. FKK-GNASH calculation of the 80 MeV ^{90}Zr(p, xp) angular distributions at proton emission energies of 20, 40, and 60 MeV, compared with experimental measurements of Cowley *et al.* [25].

Accurate modeling of intermediate-energy reactions is needed for the accelerator-base transmutation of waste (ATW), where secondary neutrons from a proton-induced reaction transmute long-lived radionuclides to stable or short-lived nuclei. To assess the modeling capabilities available, Blann *et al.* established an international code intercomparison for intermediate energy nuclear data, organized by the Nuclear Energy Agency of the OECD [17]. Participants calculated neutron and proton emission spectra from proton-induced reactions on lead and zirconium, for a range of incident energies. The majority of the codes used semiclassical preequilibrium or intranuclear cascade models. However, FKK calculations were submitted by Chadwick and Young using the FKK-GNASH code system [7], and by Koning using the KAPSIES code, and performed favorably in the intercomparison. There was evidence, though, that the FKK-GNASH calculations underpredicted backward-angle emission at the higher energies (see Fig. 4.), a problem which still needs to be solved.

The FKK-GNASH code system has also been applied to evaluate neutron and proton induced reactions on biologically-important elements. The Lawrence Livermore National Laboratory Medical Applications Program is using Monte Carlo transport to calculate dose deposition in cancer radiation treatment. Certain types of tumor respond well to neutron or proton radiation, and to simulate dose deposition, libraries of nuclear cross sections are needed. A range of modeling codes have been applied to this problem (FKK-GNASH, GNASH, ALICE) and their predictions compared with experimental measurements where available. As an example, in Fig. 5 we show the calculated inclusive proton and alpha spectrum from the n+^{12}C reaction at 60 MeV, compared with measurements from UC Davis [26]. The calculations are seen to describe the data fairly well, though the highest proton energies are underpredicted since we have not included direct mechanisms to the discrete levels. In the case of alpha emission, much of the inclusive cross section comes from the 3α breakup of ^{12}C, which we treat in a sequential way using Hauser-Feshbach theory.

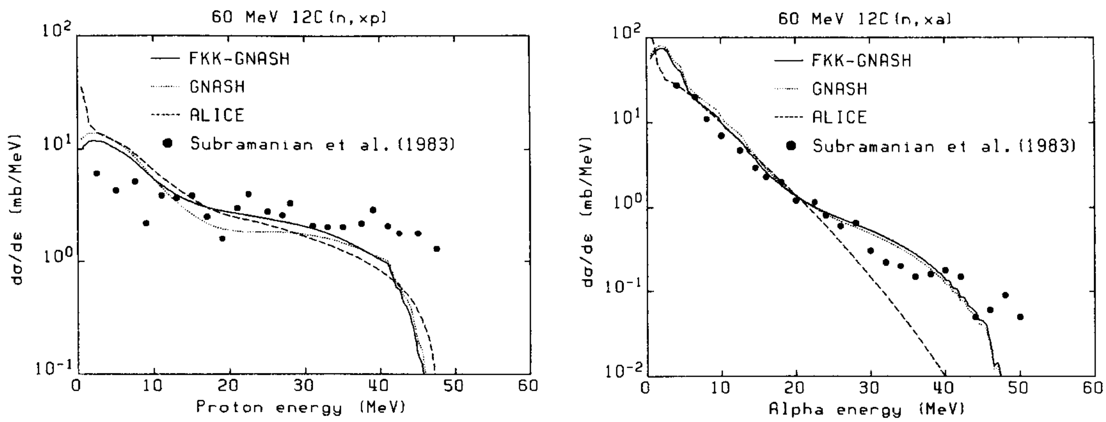

Fig. 5. Various model calculations compared with data for the 60 MeV ^{12}C(n, xp) and ^{12}C$(n, x\alpha)$ reaction.

The total kerma (*kinetic energy released in matter*) in this reaction represents an integral check on our calculations. This quantity is crucial for obtaining an accurate prediction of dose deposition in neutron therapy. In Fig. 6 we see that the FKK-GNASH and GNASH calculations describe the data well, though the omission of preequilibrium clusters in ALICE leads to an underprediction.

Finally, the FKK theory has also been used to determine radioactive isomer production in neutron-induced reactions of importance in fusion technology [4]. Particular attention was paid to spin effects in the theory. Certain reactions were selected and studied by an IAEA Coordinated Research Programme, and the evaluated cross sections at 14 MeV and at lower energies will be taken into account during the engineering design of the ITER fusion reactor.

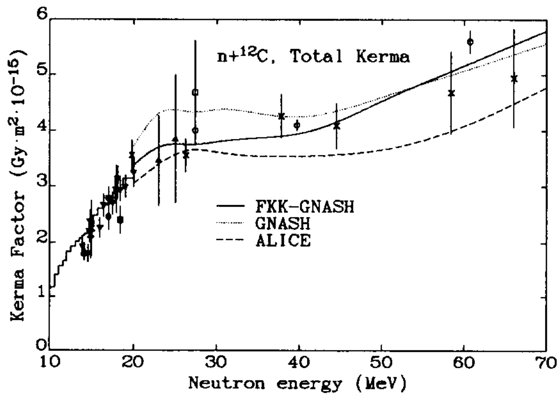

Fig. 6. Calculated total kerma factor for n+^{12}C compared with data [27].

IV. FUTURE CHALLENGES

We have given an overview of some of the recent extensions and applications of the FKK-GNASH code. Here we itemize areas which still need to be addressed:

- When a 1-component model is used, different V_0 systematics result for (p,p') and (p,n) reactions [8]. Hopefully a 2-component theory which follows neutron and proton excitations would unify these systematics.
- There is still a need for a rigorous multiple preequilibrium theory which uses basic DWBA cross sections involving more than one final continuum particle.
- Present FKK-GNASH calculations at high energies underpredict back-angle emission. Hopefully this inadequacy can be resolved. Since Koning's FKK calculations [28] do not seem to suffer from this drawback, we shall investigate the use of collective (rather than microscopic) form factors in our calculations.
- The preequilibrium emission of composite particles such as alphas and deuterons has yet to be formulated in the FKK theory. At present we use the semiclassical approach of Kalbach [29].
- Further challenges in applying the FKK theory up to 200 MeV include: use of appropriate optical potentials and level densities at high energies; relativistic effects; and inclusion of quasifree scattering physics.

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